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REVIEW ARTICLE

Malachite green "a cationic dye" and its removal from aqueous solution by adsorption

Nirav P. Raval[1] · Prapti U. Shah[1] · Nisha K. Shah[2]

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Abstract Adsorption can be efficiently employed for the removal of various toxic dyes from water and wastewater. In this article, the authors reviewed variety of adsorbents used by various researchers for the removal of malachite green (MG) dye from an aqueous environment. The main motto of this review article was to assemble the scattered available information of adsorbents used for the removal of MG to enlighten their wide potential. In addition to this, various optimal experimental conditions (solution pH, equilibrium contact time, amount of adsorbent and temperature) as well as adsorption isotherms, kinetics and thermodynamics data of different adsorbents towards MG were also analyzed and tabulated. Finally, it was concluded that the agricultural solid wastes and biosorbents such as biopolymers and biomass adsorbents have demonstrated outstanding adsorption capabilities for removal of MG dye.

Keywords Malachite green · Wastewater · Biosorbents · Biopolymers · Nanomaterials

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✉ Nirav P. Raval
nirav.raval90@gmail.com

Prapti U. Shah
prapti.shah1126@gmail.com

Nisha K. Shah
nish_chem2004@yahoo.com

[1] Department of Environmental Science, School of Sciences, Gujarat University, Ahmedabad 380 009, Gujarat, India

[2] Department of Chemistry, School of Sciences, Gujarat University, Ahmedabad 380 009, Gujarat, India

Introduction

Till the late nineteenth century, all the colorants were extracted from shells, flowers, roots, insects and molluscs. But, with the historic discovery of first synthetic dye, Mauveine, most of the uses of natural dyes have been replaced with synthetic ones as they can be manufactured on a large scale (Gordon and Gregory 1987). At present, more than 1,00,000 synthetic dyes exist with an annual production of over 7×10^5 tonnes/year and are mostly associated with water pollution (Mittal et al. 2009; Gupta et al. 2011; Saravanan et al. 2016).

Malachite green (MG) is water soluble cationic dye that appears as green crystalline powder and belongs to triphenylmethane category (Raval et al. 2016b). The important physicochemical characteristics of MG were presented in Tables 1 and Table 2 narrated its applications and toxic effects. It was revealed from the Table 2 that the extensive usage of MG dye has caused several health hazards and hence, proper treatment of effluent containing MG dye is extremely necessary.

Many treatment technologies have been applied to decolorize MG from aqueous medium, such as biodegradation and decolorization (An et al. 2002; Levin et al. 2004; Ren et al. 2006; Eichlerová et al. 2006; Daneshvar et al. 2007a, b; Ayed et al. 2008; Ali et al. 2009; Wu et al. 2009; Cheriaa and Bakhrouf 2009; Ayed et al. 2010; Du et al. 2011; Khataee et al. 2011a, b; Shedbalkar and Jadhav 2011; Hu et al. 2011; Abd-El-Kareem and Taha 2012); photocatalytic degradation (Hasnat et al. 2003; Kominami et al. 2003; Lv et al. 2005; Yang et al. 2005; Chen et al. 2007; Sayılkan et al. 2007a, b, 2008; Tayade et al. 2007; Wang et al. 2008a; Ju et al. 2008; Asiltürk et al. 2009; Prado and Costa 2009; Bansal et al. 2009; Kaneva et al. 2010; Liu et al. 2010; Bojinova and Dushkin 2011; Chen

Table 1 Physicochemical properties of malachite green dye

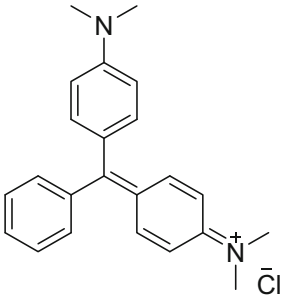
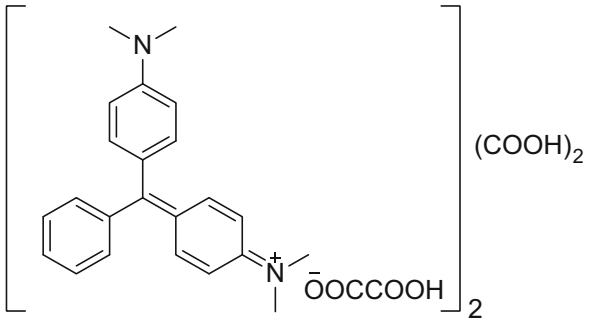
Parameters	Value	
Common name	Malachite green chloride	Malachite green oxalate
Molecular formula	$C_{23}H_{25}ClN_2$	$C_{52}H_{54}N_4O_{12}$
Molecular weight	364.911 g/mol	927.00 g/mol
IUPAC name	[4-[[4-(Dimethylamino)phenyl]-phenylmethylidene]cyclohexa-2,5-dien-1-ylidene]-dimethylazanium; chloride	[4-[[4-(Dimethylamino)phenyl]-phenylmethylidene]cyclohexa-2,5-dien-1-ylidene]-dimethylazanium;2-hydroxy-2-oxoacetate;oxalic acid
Molecular Structure		
Other names	Aniline green; Basic green 4; Diamond green B; Victoria green B	
Color index number	42000	
CAS number	123333-61-9	
Maximum wavelength	618 nm	

Table 2 Applications and toxicity of Malachite green dye

Applications
It is used for the dyeing of cotton, paper, jute, silk, wool, leather products and acrylic industries
Antiseptic and fungicidal for humans
Antiparasitical, antibacterial and antifungal in aquaculture and commercial fish hatchery industries
It is also used as a food coloring agent, food additive, a medical disinfectant, and anthelminthic
Toxicity
Environmentally persistent
Damage to nervous system, brain and liver when ingested
Eye burns, fast breathing, profuse sweating and cancer of different parts of the body
Acutely toxic to a wide range of aquatic and terrestrial animals
Decreases food intake, growth and fertility rates
Causes damage to the liver, spleen, kidney and heart
Inflicts lesions on the skin, eyes, lungs and bones
Produces teratogenic effects
Cytotoxic to mammalian cells
Acts as a respiratory enzyme poison
Decreasing RBC count (Dyscrasia), Hb (Anemia), and HTC (%)
Increasing WBC count (Leukocytosis) and delay in blood coagulation
Its presence in the hydrosphere reduces photosynthesis by obstructing light penetration and adversely affects aquatic life

et al. 2011; Tolia et al. 2012; Saha et al. 2012; Aliyan et al. 2013); photooxidative degradation (Modirshahla and Behnajady 2006); Solar degradation (Pirsaheb et al. 2015); coagulation–flocculation (Oladoja and Aliu 2009; Man et al. 2012); ozonation (Zhou et al. 2013); fenton reagent (Chen et al. 2002; Karimi et al. 2012; Bai et al. 2013; Nidheesh et al. 2013); Solvent extraction (Pandit and Basu 2004); sonochemical and sonophotocatalytic degradation (Berberidou et al. 2007; Bejarano-Pérez and Suárez-Herrera 2008; Moumeni and Hamdaoui 2012). However, all these techniques have certain limitations in terms of design, dye separation efficiency, cost and effectiveness. Advantages and disadvantages of various techniques used for the removal of MG dye have been summarized in Table 3 (Dawood and Sen 2014; Yang et al. 2014). More than 250 research articles have been published related to the adsorption of MG which confirmed that amongst all the available treatment techniques, adsorption has more frequently used one.

In addition, though there was a number of review articles such as Microbial decolorization of textile-dye containing effluents: a review by Banat et al. (1996); Fungal decolorization of dye wastewaters: a review by Fu and Viraraghavan (2001); Remediation of dyes in textile effluent: a critical review on current treatment technologies with a proposed alternative Robinson et al. (2001); The removal of color from textile wastewater using whole bacterial cells: a review by Pearce et al. (2003); Removal of synthetic dyes from wastewaters: a review by Forgacs et al. (2004); Decolourization of industrial effluents—available methods and emerging technologies—a review by Anjaneyulu et al. (2005); Adsorption–desorption characteristics of phenol and reactive dyes from aqueous solution on mesoporous activated carbon prepared from waste tires by Tanthapanichakoon et al. (2005); Non-conventional low-cost adsorbents for dye removal: a review by Crini (2006); Methods of dye removal from dye house effluent—an overview by Mondal (2008); Agricultural based activated carbons for the removal of dyes from aqueous solutions: a review by Demirbas (2009); Application of low-cost adsorbents for dye removal—a review by Gupta and Suhas (2009); Biodegradation of synthetic dyes—a review by Ali (2010); Decolorization of dye wastewaters by biosorbents: A review by Srinivasan and Viraraghavan (2010); Cationic and anionic dye adsorption by agricultural solid wastes: a comprehensive review by Salleh et al. (2011); A review on applicability of naturally available adsorbents for the removal of hazardous dyes from aqueous waste by Sharma et al. (2011); Adsorption of dyes and heavy metal ions by chitosan composites: a review by Wan Ngah et al. (2011); Microbial decolouration of azo dyes: A review by Solís et al. (2012); Adsorption of dyes using different types of sand: a review by Bello et al. (2013); Adsorptive removal of dyes from aqueous solution onto carbon nanotubes: a review by Gupta et al. (2013); Agricultural peels for dye adsorption: a review of recent literature by Anastopoulos and Kyzas (2014); Magnetic composite an environmental super adsorbent for dye sequestration—a review by Sivashankar et al. (2014); Dye and its removal from aqueous solution by adsorption: a review by Yagub et al.

Table 3 Advantages and disadvantages of various methods used for the removal of malachite green dye (Dawood and Sen 2014; Yagub et al. 2014)

Techniques	Advantages	Disadvantages
Chemical methods		
Coagulation–flocculation	Simple and economically feasible. Short detention time and low capital costs. Good removal efficiencies	High sludge production. Handling and disposal problems. High cost of chemicals for pH adjustment. Dewatering and sludge handling problems
Ozonation	Ozone can be applied in its gaseous state and does not increase the volume of wastewater and sludge	Short half–life (20 min)
Fenton's reagent	Capable of decolorizing wide variety of wastes. No attention in volume. Effective decolorization of both soluble and insoluble dyes	Prohibitively expensive. Sludge generation
Biological methods		
Biodegradation	Economically attractive. Publicly acceptable treatment	Slow process. Necessary to create an optimal favorable environment. Maintenance and nutrition requirements
Emerging technologies		
Photocatalysis	Process carried out at ambient conditions. Inputs are atoxic and inexpensive. Complete mineralization with shorter detention times. No sludge production	Effective for small amount of colorants. Expensive process. Formation of by-products
Sonication	Simplicity in use. Very effective in integrated systems	Relatively new method and awaiting full scale application

(2014), none of them has given comprehensive knowledge on the adsorptive removal of MG from aqueous media.

Therefore, in this review article we tried to congregate the available information on various adsorbents used for the removal of MG, a cationic dye, from water and wastewater. The main objective of this review article was to compile an extensive list of adsorbents which will help the future generation to furnish comprehensive up-to-date research summary on adsorbents used for removal of MG dye. Also the reported optimal experimental conditions (solution pH, contact time and amount of adsorbent) as well as adsorption isotherms, kinetics and thermodynamics data of different adsorbents towards MG were also mentioned. The authors embolden the readers to refer to the original research articles for information regarding the experimental conditions.

Dyes and their classification

Dyes (a natural or synthetic substance) are mainly chemical compounds that can affix themselves on the surface of fabrics to impart color (Yagub et al. 2014). It refers as a substance, usually organic, which is designed to be absorbed or adsorbed by, made to react with, or deposited within a substrate to impart color to that substrate with some degree of permanence (Burkinshaw 2016). Dyes may be classified in several ways (Adegoke and Bello 2015). Figure 1 summarizes the complete classification of dyes.

Based on the source of materials: Dyes can be classified mainly into two types based on their origin

Natural dyes

Natural dyes can be derived from the three primary sources—plants, animals and minerals. They are mostly negatively charged. Positively charged natural dyes can also exist, but they are not very common. Irrespective of the source of origin, natural dyes can be categorized as:

1. Substantive dyes: Those dyes which become chemically fixed to the fiber without the aid of any other chemicals or additives are referred to as substantive dyes or direct dyes.
2. Adjective dyes: Those natural dyes which require an added substance or mordant to make the dyes colorfast are known as adjective dyes or mordant dyes. Most of the natural dyes belong to this category (Cardon 2007).

Synthetic dyes

Dyes derived from organic or inorganic compounds are known as synthetic dyes. Because of their easy use, cost effectiveness and wide range of colors these dyes became popular.

Based on their method of application to the substrate

Acid dyes

Acid dyes are highly water soluble anionic dyes which contain one or more sulfonic groups/substituents and other acidic groups. They have better light fastness compare to basic dyes. The existence of sulfonic acid groups upsurge their solubility in water and give the dye molecules a negative charge. Acid dyes can be further divided into two sub-groups:

1. Acid-leveling dyes: These dyes are smaller or medium sized, and show a moderate affinity for wool fibers. Hence, due to average affinity these dyes are not very resistant to washing.
2. Acid-milling dyes: These dyes are larger than acid-leveling dyes, and show a much stronger intermolecular attractions for wool fibers. Due to this strong affinity these dyes are very resistant to washing.

Basic dyes

These water soluble dyes possess cationic functional groups such as $-NR^{3+}$ or $=NR^{2+}$. They are generally amino and substituted amino compounds soluble in acid. They become attached to the fibers by formation of ionic bonds with fiber's anionic groups.

Direct dyes

Although these dyes are water soluble anionic dyes, they cannot be classified as acid dyes because the acid groups are not the means of attachment to the fiber. Since, these dyes do not require any kind of fixing, they are called direct dyes.

Mordant dyes

A latin word mordant means 'to bite'. It acts as 'fixing agent' to improve the color fastness of some acid dyes, because they have the ability to form insoluble colored complexes with metal ions. They are usually metal salts. Besides, alum there is a large range of other metallic salt mordants available. Aluminum, chromium and iron salts are frequently used for synthetic mordant dyes.

Reactive dyes

Fiber reactive dyes will form a covalent bond while they react with the cellulosic fiber. As a result they are very

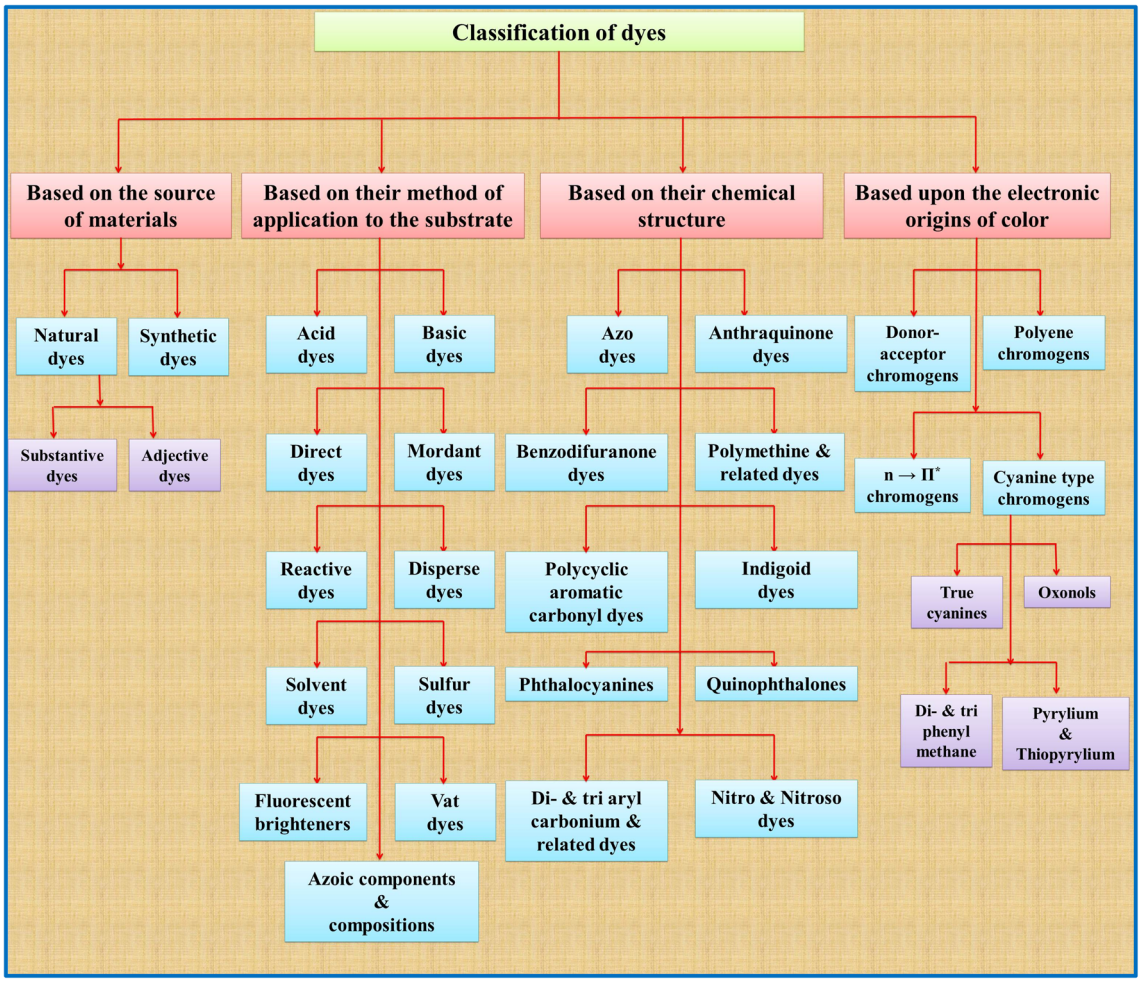


Fig. 1 Broad classification of dyes

difficult to remove, once attached with the fiber. Initially, these dyes were designed for cellulose fibers but now some fiber-reactive dyes for protein and polyamide fibers are also commercially available.

Disperse dyes

These dyes have substantially low solubility in water. Their structure is small, planner and non-ionic with attached polar functional groups, such as $–NO_2$ and –CN. They are mainly used for the dyeing of polyesters because they can interact with the polyester chains by forming dispersed particles.

Solvent dyes

These dyes are insoluble in water but soluble in alcohols, chlorinated hydrocarbons and liquid ammonia. They are mainly used for coloring plastics, synthetic fibers, gasoline, oils and waxes.

Sulfur dyes

Sulfur dyes are applied to cotton from an alkaline reducing bath with sodium sulfide as the reducing agent. These dyes are water insoluble but they are soluble in their reduced form and exhibit affinity for cellulose. They are low cost and have good fastness to light, washings and acids. The actual structures of sulfur dyes are largely unknown although it is considered that they possess sulfur-containing heterocyclic rings.

Based on their chemical structure: (Gordon and Gregory 1987; Waring and Hallas 1990)

Azo dyes

Approximately half of all dyes are azo dyes, making them the largest group of synthetic colorants used in textile industries. This type of dyes contain minimum one azo group but can also contain two or three, but very rarely,

four azo groups. Azo dyes are complex aromatic compounds with significant structural diversity and are of great environmental concern because the reductive cleavage of azo linkages is responsible for the formation of amines, which are classified as toxic and carcinogenic (Raval et al. 2016a).

Anthraquinone dyes

Anthraquinone dyes are the second most important class after azo dyes. They are also one of the oldest types of dyes since they have been found in the covering of mummies dating back over 4000 years. In contrast to the azo dyes, which have no natural counterparts, all the natural red dyes were anthraquinones. The lower tinctorial strength and reduced flexibility increase the production cost of anthraquinone dyes and hence, they are not as widely used as azo dyes.

Benzodifuranone dyes

Benzodifuranone (BDF) dyes are one such type which attacking anthraquinone dyes. The BDF chromogen is one of the very few novel chromogens to have been accidently discovered this century

Polymethine and related dyes

Dyes containing a conjugated chain of carbon atoms terminated by an ammonium group and, in addition, a nitrogen, sulfur, or oxygen atom, or an equivalent unsaturated group is termed as polymethine dyes. This type of dyes may be neutral, cationic or anionic. The best known cationic polymethine dyes are the cyanine dyes. Cyanine dyes cannot be used as textile dyes because they have poor light fastness. They are used in photography.

Polycyclic aromatic carbonyl dyes

Polycyclic aromatic carbonyl dyes have one or more carbonyl groups. These carbonyl groups are linked by a quinonoid system. These dyes are relatively very large molecules which are made up from smaller units of anthraquinones. These dyes are often termed as anthraquinonoid vat dyes because they are applied to the substrate by a vatting process.

Indigoid dyes

Indigoid dyes also contain carbonyl groups, similar to the anthraquinone, benzodifuranone and polycyclic aromatic carbonyl dyes. They are vat dyes and represent one of the oldest known classes of dyes.

Phthalocyanines

Phthalocyanines are one of the novel chromophores which have been discovered since the nineteenth century. They are structurally similar to the natural porphyrin pigments haemin and chlorophyll. These natural pigments have poor stability, but phthalocyanines have exceptional stability and tinctorially stronger. All the phthalocyanine compounds are blue to green in color because substituents in their fused benzene rings exert only a minor sway on the color of phthalocyanines. They are widely used in printing inks and paints.

Di- and tri-aryl carbonium and related dyes

Di- and triarylcarbonium dyes and their heterocyclic derivatives comprise the oldest class of synthetic dyes—the majority of which were discovered in the 19th and early 20th century. Diphenylmethane and Triphenylmethane dyes are the most important aryl carbonium dyes. Structurally and property wise they are similar to the cyanine dyes i.e. exceptional brightness, high tinctorial strength and low light fastness.

Nitro and nitroso dyes

Nitro and nitroso dyes are minor commercial importance and constitute only a fraction of the total dyes market. Nitro dyes have simple chemical structures. They are small aromatic molecules which contain minimum one nitro group and one or more amino or hydroxy groups. Initially, the nitro dyes were acid dyes which were used for dyeing the natural animal fibers i.e. wool and silk. Picric acid was the first synthetic nitro dye which imparted a greenish-yellow color to silk but could not be used for longer period of time because of its toxicity and poor fastness properties.

Nitroso dyes are prepared by nitrosation of the appropriate phenol or naphthol. They are generally metal complex derivatives of *o*-nitrosophenols or naphthols. Nitroso dyes are polygenetic because they form differently colored complexes with different metals. They have good light fastness.

Based upon the electronic origins of color

Griffith (1976, 1981) has classified dyes based on the electronic mechanism by which color is generated. According to this, purely on theoretical basis, dyes can be arranged into four classes.

(II) Donor–acceptor chromogens
(III) Polyene chromogens
(IIII) $n \rightarrow \pi^2$ chromogens

(IIV) Cyanine type chromogens

Literature on various adsorbents used for the removal of MG

See (Fig. 2)

Activated carbon adsorbents

Activated carbons (AC) are known as very efficacious adsorbents due to their large surface area, a micro-porous structure and a high degree of surface reactivity. However, due to their high production costs, these materials tend to be more expensive than other adsorbents. Four classes of AC include: powder (PAC), granular (GAC), fibrous (ACF) and cloth (ACC). Also two more frequently used ACs are: (1) commercial activated carbons and (2) activated carbons prepared from agricultural waste materials (Raval et al. 2016c).

Commercial activated carbons

A number of commercially available activated carbons such as powdered activated carbon (Kumar 2006); coal-based activated carbon (Aitcheson et al. 2000); commercially available powdered activated carbon (CPAC) (Malik et al. 2007); activated carbon prepared from lignite (Önal et al. 2007); activated Charcoal (Iqbal and Ashiq 2007); activated carbon prepared from Tuncbileklignite (Önal et al. 2006); oxyhumolite (Young brown coal) (Janos et al. 2005); Ordered mesoporous carbons (OMCs) (Tian et al. 2011); etc. have been extensively studied for the adsorption of MG.

Mesoporous carbon was synthesized by Anbia and Ghaffari (2012) for the removal of a cationic dye malachite green (MG) from aqueous solution. They carried out study under various experimental conditions and concluded that the equilibrium was attained within 30 min of contact with adsorbent and more than 99% removal of MG was reached at the optimum pH value of 8.5.

Aitcheson et al. (2000) used coal-based activated carbon 207EA (Sutcliffe Speakman) for the removal of aquaculture therapeutants (Malachite Green, formalin, Chloramine-T and Oxytetracycline) which were widely used in aquaculture to control fish parasites and disease. The adsorption process was well fitted to the Freundlich isotherm and confirmed that adsorption was heterogeneous in nature.

Adsorption of MG was studied by Mall et al. (2005) using commercial grade (ACC) and laboratory grade (ACL) activated carbons. Authors conducted batch adsorption studies to evaluate the effect of various parameters such as pH, adsorbent dose, contact time and initial MG concentration on the removal of MG and concluded that the effective pH was 7.0 for adsorption of MG. Equilibrium reached in about 4 h contact time and optimum ACC and ACL dosages were 20 and 4.0 g/L, respectively.

To overcome deficiencies of CACs the use of activated carbon adsorbents derived from various agricultural waste and industrial solid waste materials have received increasing consideration in the purification of wastewater over the last several years because they are generated in excess, inexpensive, locally available and their use will

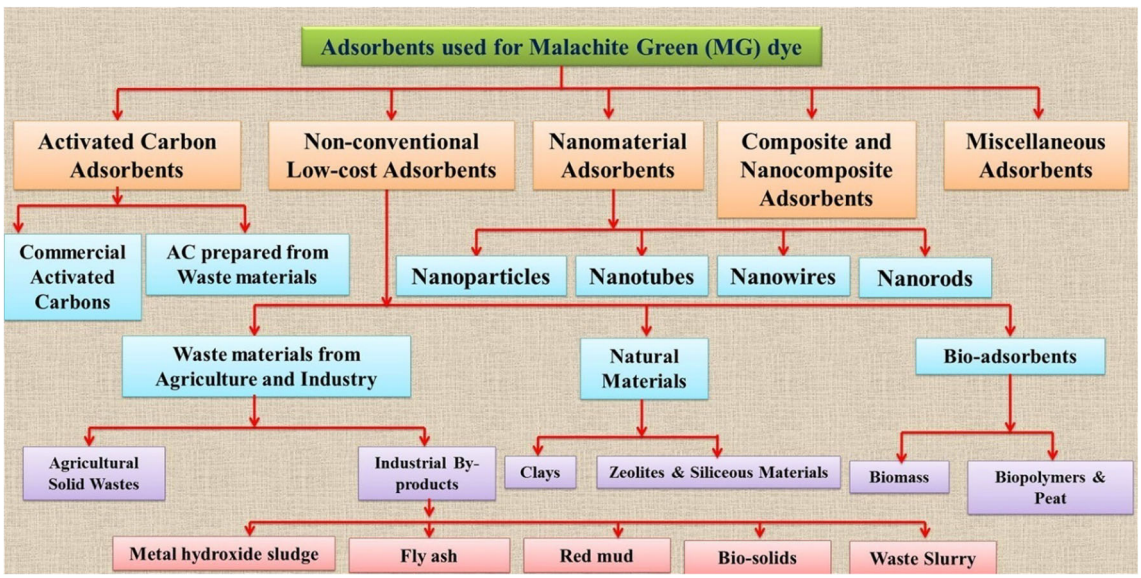


Fig. 2 Classification of adsorbents used for the removal of MG

further solve the problem of their disposal (Pehlivan and Arslan 2007).

Activated carbons prepared from agricultural and industrial solid wastes

As biomass and other waste materials have little or no economic value and often present a disposal problem, they offer an inexpensive and additional renewable source of activated carbon. Thus, conversion of these low cost by products into activated carbon would provide an inexpensive substitute to the existing CACs, add monetary value and help in dropping the expenditure of waste disposal (Raval et al. 2016c).

Polygonum orientale Linn is an annual plant of the genus *Polygonum* in the family Polygonaceae, which is native to eastern Asia and widely distributed in China. This plant has a developed caudex system and generally produces large and continuous caudex masses. Therefore, this plant offers a good basis for the development of adsorbent materials. Looking upon this property of plant (Wang et al. 2010) prepared *P. orientale* Linn activated carbon (PLAC) from waste *P. orientale* Linn by chemical activation with H_3PO_4 and used as an adsorbent to remove malachite green (MG) and rhodamine B (RB) from aqueous solution.

Durian (*Durio zibethinus* Murray) is the most popular seasonal fruit in Southeast Asia, particularly Malaysia, Indonesia, Thailand, and the Philippines, and belongs to the family Bombacaceae. A significant percentage of the planted durian fruit crop is wasted each year. Only one third of durian fruit is edible, whereas the seeds (20–25%) and the shells are usually discarded. The wasted durian seed represents a significant potential for the development of value-added products. Thus, Ahmad et al. (2014) studied the removal of Malachite green dye using durian seed-based activated carbon.

Rambutan (*Nephelium lappaceum* L.) is a popular tropical fruit which belongs to Sapindaceae family. However, due to the high consumption of rambutan's edible part, massive amount of the peel is disposed, causing a severe problem in the community as they gradually ferment and release off odors. Therefore, Ahmad and Alrozi (2011) converted rambutan peel into activated carbon by physiochemical activation for its potential to remove MG dye from wastewater.

Akar et al. (2013) produced active carbon from spent tea leaves (STAC) by activation with 4% NaOH solution for the removal of malachite green dye because tea is basically dried and processed leaves of plant called *Camellia sinensis*. It is consumed by the most of people in the world and is the second most popular beverage in the world. It is estimated that between 18 and 20 billion cups of tea are drunk daily in world and disposal of spent leaves become a major problem.

Banana (*Musa paradisiaca*) is native to Southeast Asia, most of the bananas produced are consumed locally and ~10% are exported. Massive amounts of stalk (as waste products) are disposed, resulting in a serious environmental problem through the emission of foul odors when it is left to decompose. Hence, Bello et al. (2012) attempted to use banana stalk, an abundantly available lignocellulosic agricultural waste, as a cheap and renewable precursor for activated carbon preparation for MG dye removal and helped in placing value on this agricultural waste and provide a potentially cheap alternative to existing commercial activated carbons.

In addition to above described materials, a wide variety of other agricultural and industrial waste materials have also been used for the preparation of activated carbon and applied for the adsorptive removal of MG dye from aqueous solution. It includes Tamarind fruit shells carbon (Vasu 2008); *Juglans regia* shells carbon (Nethaji et al. 2012); *Annona squmosa* seed (Santhi et al. 2010a); Groundnut shell carbon (Malik et al. 2007); Pine sawdust (Akmil-Başar et al. 2005); Tea leaves (Singh and Rastogi 2004); Bamboo (Hameed and El-Khaiary 2008a); Jute fiber (Porkodi and Vasanth Kumar 2007) and waste printed circuit boards (Kan et al. 2015), etc.

In addition, Table 4 presented various commercially available and agricultural waste based activated carbon adsorbents used for the removal of MG and their optimum conditions.

Non-conventional low-cost adsorbents

Activated carbon adsorption has been cited by the US Environmental Protection Agency as one of the best available control technologies. However, although activated carbon is a preferred sorbent, its widespread use is restricted due to high cost. To decrease the cost of treatment, attempts have been made to find inexpensive alternative adsorbents. Recently, numerous approaches have been studied for the development of cheaper and effective adsorbents. Thus, there is a need to search for new and economical adsorbent that could remove metal. In this context, agricultural by-products and industrial waste can be seen as having a great potential to be developed as a low cost sorbent. A low-cost adsorbent is defined as the one which is plentiful in nature, or is a by-product or waste from industry and requires little or no processing (Raval et al. 2016c). Literature survey reveals that multifarious materials have been utilized as adsorbents under this category.

Table 4 The maximum adsorption capacities and other optimal conditions of commercially available and agricultural waste based activated carbon adsorbents for adsorption of MG from water and wastewater

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
1.	Powdered activated carbon	509.00	Langmuir and Redlich–Peterson	–	Exothermic	–	–	–	(Kumar 2006)
2.	Activated carbon	490.77	Langmuir and Redlich–Peterson	Pseudo 2nd order	–	–	–	–	(Kumar and Sivanesan 2006)
3.	Mesoporous Carbon	476.10	Langmuir	Pseudo 1st order	–	8.5	30	0.005 g/25 mL	(Anbia and Ghaffari 2012)
4.	Coal-based activated carbon	325.00	Freundlich	–	–	–	–	–	(Aitcheson et al. 2000)
5.	Commercially available powdered activated carbon (CPAC)	222.22	Freundlich	–	–	–	15	0.7 g/L	(Malik et al. 2007)
6.	Activated carbon prepared from lignite	200.00	Langmuir	Pseudo 2nd order	Endothermic	7.0	60 min	0.1 g/50 mL	(Önal et al. 2007)
7.	Activated Carbon	75.08 (batch capacity) 94.00 (column capacity)	Langmuir and Freundlich	Pseudo 1st order	Endothermic	10.0	6–8 h	1.0 g/L	(Gupta et al. 1997)
9.	Laboratory grade activated carbon (ACL)	42.18	Redlich–Peterson	Pseudo 2nd order	–	7.0	240	4 g/L	(Mall et al. 2005)
10.	Acid-activated carbon	9.78	Langmuir	–	Endothermic	6.0	40 min	–	(Hema and Arivoli 2007)
11.	Commercial grade activated carbon (ACC)	8.27	Freundlich	Pseudo 2nd order	–	7.0	240	20 g/L	(Mall et al. 2005)
12.	Activated Charcoal	0.180	Langmuir	–	Exothermic	7.0	30 min	0.1 g	(Iqbal and Ashiq 2007)
13.	Activatedcarbon prepared from Tunc¸bileklignite	9.92×10^{-4} (mol/g)	Freundlich	Pseudo 2nd order	Endothermic	5.0	120 min	0.1 g/25 mL	(Önal et al. 2006)
14.	Oxyhumolite (Young brown coal)	0.278 mmol/g	Langmuir	–	–	5.0	–	–	(Janos et al. 2005)
15.	Activated carbon	26.19	Langmuir	Pseudo 2nd order	Endothermic	7.0	60 min	0.5 g/50 mL	(Arivoli et al. 2008)
16.	Oxyhumolite (Young brown coal)	0.278 mmol/g	Langmuir	–	–	–	–	–	(Janoš et al. 2007)
17.	Ordered mesoporous carbons (OMCs)	–	Freundlich	Pseudo 2nd order	Exothermic	–	–	0.07 g/25 mL	(Tian et al. 2011)
18.	*Polygonum orientale* Linn activated carbon (PLAC)	556.00	Langmuir	Pseudo 2nd order	Endothermic	10.0	150 min	0.45 g/L	(Wang et al. 2010)
19.	Durian seed activated carbon (DSAC)	476.19	Freundlich	Pseudo 2nd order	Endothermic	8.0	24 h	0.1 g	(Ahmad et al. 2014)
20.	Pine sawdust by chemical activation with $ZnCl_2$	370.37	Langmuir	Pseudo 1st order	–	10.0	120 min	0.1 g/25 mL	(Akmil-Başar et al. 2005)

Table 4 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
21.	Rambutan peel-based activated carbon (RPAC)	329.49	Freundlich	Pseudo 2nd order	Endothermic	8.0	24 h	0.2 g	(Ahmad and Alrozi 2011)
22.	Tea leaves based activated carbon	275.00	Langmuir and Freundlich			8.0–10.0		1.2 g/L	(Singh and Rastogi 2004)
23.	Bamboo-based activated carbon	263.58	Langmuir	Pseudo 2nd order	–	4.0	170 min	0.2 g	(Hameed and El-Khaiary 2008a)
24.	Spent tea leaves active carbon (STAC)	256.40	Langmuir	Pseudo 2nd order	Endothermic	4.0		0.8 g/L	(Akar et al. 2013)
25.	Groundnut shell based powdered activated carbon (GSPAC)	222.22	Freundlich	–	–	–	30	0.7 g/L	(Malik et al. 2007)
26.	Cattail carbon	196.08	–	–	–	–	–	–	(Shi et al. 2010)
27.	Activated carbon prepared waste apricot	163.93	Langmuir and Frumkin	–	Endothermic	–	60 min	0.1 g/ 50 mL	(Başar 2006)
28.	Banana (*Musa paradisiaca*) stalk-based activated carbon	141.76	Langmuir	Pseudo 2nd order	Endothermic	8.0	120 min	0.2 g	(Bello et al. 2012)
29.	Jute fiber carbon (JFC)	136.58	Freundlich	Pseudo 2nd order	–	8.0	220 min	0.05 g	(Porkodi and Vasanth Kumar 2007)
30.	Activated carbon prepared waste apricot	111.60	–	Pseudo 2nd order	Endothermic	–	60 min	0.1 g/ 50 mL	(Onal 2006)
31.	Tamarind fruit shells activated carbon	83.41	Langmuir	Pseudo 2nd order	Endothermic	8.0			(Vasu 2008)
32.	Rubber seed coat based activated carbon (RSCAC)	72.73	Langmuir	–	Endothermic	6.0	3 h		(Idris et al. 2011)
33.	Rice husk activated carbon	63.85	Langmuir	Pseudo 2nd order	Exothermic		40 min		(Sharma et al. 2009)
34.	Activated carbon derived from *Borassus aethiopum* flower biomass (PFAC)	48.48	Langmuir	Pseudo 2nd order	Endothermic	6.0–8.0	24 h	0.1 g/ 10 mL	(Nethaji et al. 2010)
35.	Rice husk activated carbon (RHAC)	49.62	Langmuir	Pseudo 2nd order	Endothermic	8.0	40 min		(Sharma 2011)
36.	Water nut modified carbon (WNMC)	46.27	Langmuir	Pseudo 2nd order	Endothermic	8.0		10 mg/ 50 mL	(Ahmad and Mondal 2010)
37.	*Cucumis sativa* fruit peel based activated carbon	36.230	Langmuir	Pseudo 2nd order	–	6.0	50 min	1 g/50 mL	(Santhi and Manonmani 2011)
38.	*Juglans regia* shells-based activated carbon	29.74	Freundlich	Intraparticle diffusion and Boyd models	Endothermic	6.0–7.0	–	3.0 g/L	(Nethaji et al. 2012)
39.	Activated carbon prepared from the epicarp of *Ricinus communis*	27.78	Langmuir	Pseudo 2nd order	–	7.0	50 min	1 g/50 mL	(Santhi et al. 2010b)
40.	Coconut coir activated carbon	27.44	Langmuir	Pseudo 2nd order	Endothermic	–	–	–	(Uma et al. 2013)
41.	*Annona squmosa* seed based activated carbon	25.91	Temkin	Pseudo 2nd order	–	7.0	–	–	(Santhi et al. 2010a)
42.	Arundo donax root carbon (ADRC)	9.35	Langmuir	Pseudo 2nd order	Endothermic	5.0	180 min	0.6 g/ 100 mL	(Zhang et al. 2008)

Table 4 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
43.	Cassava (*Manihot esculenta*) peels based carbon	–	–	–	–	–	–	–	(Rajeshwarisivaraj et al. 2001)
44.	Rubber wood (*Hevea brasiliensis*) sawdust carbon	–	Langmuir	Pseudo 2nd order	Endothermic	–	3.0 h	100 mg	(Rajgopal et al. 2006)
45.	Rice husk-based porous carbon	–	Freundlich	–	–	–	–	–	(Guo et al. 2003b)
46.	*Ulva lactuca* and *Systoceira stricta* algae-based activated carbons	–	Langmuir	Pseudo 2nd order	Endothermic	4.0	90 min	10 g/L	(Salima et al. 2013)
47.	Rice husk-based porous carbons (RHCs)	–	Freundlich	–	Endothermic	6.59	90 min	0.8 g/L	(Guo et al. 2003a)
48.	Activated *Enteromorpha* carbon	–	Langmuir	Pseudo 2nd order	–	4.0	3 h	250 mg	(Jayaraj et al. 2011)
49.	Activated carbons prepared from the agricultural solid wastes	–	–	–	–	–	–	–	(Kadirvelu et al. 2003)
50.	Activated parthenium carbon	–	–	–	–	6.35	30 min	–	(Rajeshwarisivaraj and Subburam 2002)

Waste materials from agriculture and industry

Agricultural solid wastes Agricultural solid wastes are relatively cheap and are available in huge quantities. They can be used as an adsorbent due to their physic-chemical properties. The utilization of these wastes can play a significant role in national economy (Crini 2006).

Orange peel is discarded in the orange-juice and soft-drink industries all over the world. It has been used as an adsorbent for the removal of MG from wastewater by Kumar and Porkodi (2007). The results revealed that the adsorption process followed both Langmuir and Redlich–Peterson isotherms, the adsorption capacity was 483.63 mg/g.

A study of Rahman et al. (2005) reported the removal of MG from aqueous solutions onto Phosphoric acid (H_3PO_4) and sodium hydroxide (NaOH) treated rice husks.

Rice straw is a lignocellulosic agricultural stalk containing cellulose (37.4%), hemi-cellulose (44.9%), lignin (4.9%) and silica ash (13.1%). Gong et al. (2006) compared MG adsorption efficiency of native and thermos chemically modified rice straw and found their adsorption efficiency 94.34 and 256.41 mg/g, respectively.

Chieng et al. (2015) investigated the use of breadnut (*Artocarpus camansi*) as a low-cost adsorbent for the removal of MG toxic dye. In Brunei Darussalam, breadnut was locally known as '*Kemangsi*', whose seeds were consumed either cooked or roasted. The skin and core of *Artocorpus* fruits, the major parts of the fruits, were inedible and have no economical value, and discarded as waste. Authors concluded that the adsorption characteristics of breadnut skin towards MG can be significantly enhanced by modification through treatment with dilute NaOH solution.

Other solid wastes from cheap and readily available resources have also been successfully employed for the removal of MG from aqueous solution and were given in Table 5.

Industrial by-products/industrial solid wastes Industrial by products, such as metal hydroxide sludge, fly ash, red mud and bio-solids, can be used as low-cost adsorbents for MG removal. Table 6 represented the maximum adsorption capacities together with the isotherm, kinetics and thermodynamic results of various industrial solid wastes adsorbents for MG.

Fly ash: Witek-Krowiak et al. (2012) investigated the use of fly ash for the adsorption MG from solution with different contact times, initial dye concentrations, pHs and temperatures. The adsorption equilibrium has been described in terms of Langmuir model. The enthalpy values obtained in the experiment were not high, which suggested

that physical sorption dominates in the process of cationic dyes bonding on microspheres of fly ash.

Bagasse fly ash (BFA) is a waste collected from the particulate separation equipment attached to the flue gas line of the sugarcane bagasse-fired boilers and has been used as an effective adsorbent for the removal of dyes. This is available in plenty, and almost free of cost from the sugar mills. In addition, bagasse fly ash generated in the sugar industry does not contain large amounts of toxic metals and hence, Mall et al. (2005) used bagasse fly ash (BFA) as adsorbent for the removal of MG from aqueous solutions. Maximum adsorption capacity for MG was reported as 170.33 mg/g (at pH 7.0 and adsorbent dosage of 1.0 g/L) and within 240 min of contact time.

Waste red mud: Zhang et al. (2014b) used the acid activated (Acid treatment neutralize the alkalinity of red mud, which could also be recognized as a method for activating the material) sintering process red mud (ASRM) for the adsorption of MG. The optimum process variables were found to be pH $>$ 3.2, 180 min of contact time and 0.25 g/250 mL of adsorbent dosage. The adsorption process followed the Langmuir isotherm with 336.40 mg/g of adsorption capacity.

Activated slag: Steel plants produce granular blast furnace slag as a byproduct, and this material also causes a disposal problem. Hence, Gupta et al. (1997) utilized this waste product for the adsorption of MG from aqueous medium both in batch and column modes. Further, they concluded that the column capacity was found to be higher than the batch capacity. The removal takes place through a film diffusion mechanism at lower concentrations and by particle diffusion at higher concentrations.

Natural materials

Clays Clay is mineral comprised of alumina and silica that usually includes bound water. Clays have a sheet-like structure and are composed of tetrahedrally arranged silicates and octahedrally arranged aluminates. Due to their low-cost, abundance in nature, high sorption capacity, chemical and mechanical stability, layered structure and high cation-exchange capacity (CEC), clay minerals are frequently used as an adsorbent (Krauskopf 1956; da Fonseca et al. 2006).The clays always contain exchangeable ions on their surface and play vital role in the environment by acting as a natural scavengers of contaminants by taking up cations and/or anions through either ion exchange or adsorption (Babel and Kurniawan 2003).

A low-cost inorganic powder (Persian Kaolin) was used as adsorbent for the sequestration of cationic dye from aqueous solution by Tehrani-Bagha et al. (2011). The sorption of the dye malachite green on kaolin confirmed to linear form of Langmuir adsorption equation. The maximum adsorption capacity was 52.91 mg/g. In addition, kinetic and thermodynamic parameters, such as enthalpy (ΔH) and free energy (ΔG) were calculated. The negative values of ΔG and positive values of ΔH indicated that the dye adsorption process was spontaneous and endothermic in nature

The efficacy of montmorillonite clay had been investigated by Tahir et al. (2010) for the adsorptive removal of malachite green dye from aqueous solution. Authors concluded that the MG dye was homogenously adsorbed on the surface of montmorillonite clay adsorbent.

Bulut et al. (2008) investigated the sorption of MG from aqueous solution onto bentonite as a function of contact time, pH and initial dye concentration. The adsorption equilibriums have been described in terms of Langmuir, Freundlich, Temkin and Dubinin–Radushkevich models. Authors reported that the adsorption process obeyed the Langmuir isotherm and pseudo-second-order kinetic model. The results of the thermodynamic study indicated that the sorption process was endothermic and spontaneous.

Tahir and Rauf (2006) utilized the natural bentonite clay for the adsorption of MG dye from aqueous solution. The authors finally concluded that the values of sorption free energy (E_a) of 1.00–1.12 kJ/mol indicated physical adsorption of dye on clay mineral.

Siliceous materials Siliceous rocks are sedimentary rocks that have silica (SiO_2) as the principal constituent. The most common siliceous rock is chert other types include diatomite. They commonly form silica-secreting organisms, such as radiolarians, diatoms, or some types of sponges. The natural siliceous materials used for MG adsorption were perlite, silica, glass fibers and alunite because of their high abundance, easy availability and low cost.

Govindasamy et al. (2009) studied the removal of MG from aqueous solution by perlite. Authors reported 40 min of contact time, 0.1 g/50 mL of adsorbent dosage and 8.0–9.0 pH value were optimum conditions for the removal of dye. It was suggested that the adsorption of MG onto perlite was spontaneous, chemical and exothermic in nature and followed Freundlich isotherm as well as pseudo-first-order kinetic rate model.

Silica gel is a hydrophilic porous adsorbent. Aromatic compounds are found to involve the pi-cloud in hydrogen bonding with silanol (SiOH) group during adsorption. Cationic and nonionic surfactants adsorb on silica surface involving hydrogen bonding (Parida et al. 2006).

Kannan et al. (2008) utilized tetrahedral silica for the adsorption of MG. The adsorbent was characterized by Fourier transformed infrared spectroscopy (FT-IR) to confirm the tetrahedral framework of silica. The

Table 5 The maximum adsorption capacities and other optimal conditions of agricultural solid wastes for adsorption of MG from water and wastewater

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
1.	Orange Peel	483.63	Langmuir and Redlich–Peterson	–	–	–	–	–	(Kumar and Porkodi 2007)
2.	Pyromellitic dianhydride modified sugarcane bagasse	377.40	Langmuir	Pseudo 2nd order	–	6.0	13 h		(Yu et al. 2011)
3.	NaOH-modified breadnut peel	353.00	Sips	Pseudo 2nd order	Exothermic	8.02	240 min	–	(Chieng et al. 2015)
4.	NaOH-modified grapefruit peel (MGP)	314.90	Koble-Corrigan and Dubinin-Radushkevich models	Elovich model	Endothermic	–	–	–	(Zou et al. 2013)
5.	Oxalic acid modified *Cinnamomum camphora* sawdust (OACS)	282.50	Langmuir	Pseudo 2nd order	Endothermic	8.0	3 h	1.0 g/L	(Wang et al. 2014a)
6.	Modified rice straw (MRS) [rice straw was modified with citric acid (CA)]	256.41	Langmuir and Freundlich	Pseudo 1st order	–	6.0	10 h	2.0 g/L	(Gong et al. 2006)
7.	Citric acid modified *Cinnamomum camphora* sawdust (CACS)	260.40	Langmuir	Pseudo 2nd order	Endothermic	8.0	3 h	1.0 g/L	(Wang et al. 2014a)
9.	Eggshells	243.20	Freundlich	–	–	6.0	–	1.0 g/L	(Podstawczyk et al. 2014)
10.	Functionalized sawdust (FS)	196.08	Langmuir	–	Exothermic	6.0	4.5 h	2.0 g/L	(Gong et al. 2009)
11.	Treated ginger waste (TGW)	188.60	Langmuir	Pseudo 2nd order	Endothermic	9.0	150 min	0.5 g	(Ahmad and Kumar 2010)
12.	Breadnut peel	180.00	Freundlich	Pseudo 2nd order	Endothermic	8.02	240 min (2 h)	–	(Chieng et al. 2015)
13.	Phosphoric acid esterifying soybean hull	178.57	Langmuir	Pseudo 2nd order	Exothermic	6.0	–	2.0 g/L	(Gong et al. 2008)
14.	Pomelo (*Citrus grandis*) peels	178.43	Langmuir	Pseudo 2nd order	Endothermic	8.0	240 min	0.2 g	(Bello et al. 2014)
15.	Treated jack fruit peel	166.37							(Inbaraj and Sulochana 2002)
16.	Ethylenediaminetetraacetic dianhydride (EDTAD) modified sugarcane bagasse (SB)	157.20	Langmuir	Pseudo 2nd order	Endothermic	6.0	3 h	10 mg	(Xing and Deng 2009)
17.	Tartaric acid modified *Cinnamomum camphora* sawdust (TACS)	156.70	Langmuir	Pseudo 2nd order	Endothermic	8.0	3 h	1.0 g/L	(Wang et al. 2014a)
18.	Oil palm trunk fiber (OPTF)	149.35	Langmuir	Pseudo 1st order	–	8.0	195 min	0.30 g	(Hameed and El-Khaiary 2008b)
19.	Rice straw-derived char (RSC)	148.74	Langmuir	Pseudo 1st order	–	3.0–7.0	–	1.5 g/L	(Hameed and El-Khaiary 2008c)
20.	Effective Microorganisms based compost (EMKC)	136.60	Sips	Pseudo 2nd order	–	8.0	–	4.0 g/L	(Bhagavathi Pushpa et al. 2015)

Table 5 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
21.	Neem leaf powder	133.6	Langmuir	Pseudo 1st order	Endothermic	–	300 min	0.63 g/L	(Bhattacharyya and Sarma 2003)
22.	Formaldehyde-treated *Simarouba glauca* seed shell powder	125.00	Langmuir	Pseudo 2nd order		8.0	60 min	0.15 g	(Jeyagowri and Yamuna 2015)
23.	Cellulose modified with phthalic (P) anhydride (CPA)	111.00	Langmuir	Pseudo 2nd order	Endothermic				(Zhou et al. 2015)
24.	Poly(methacrylic acid)-modified sugarcane bagasse	103.20	Langmuir	Pseudo 2nd order		6.0	3 h		(Xing and Wang 2009)
25.	Brinjal plant root powder (cellulose)	100.29	Freundlich	Pseudo 2nd order	Endothermic	6.0	25 min	250 mg/ 50 mL	(Buvaneswari and Kannan 2011)
26.	Oxidized-activated form HTC-APN	97.08	Langmuir	Pseudo 2nd order	Endothermic	7.0	–	2.0 g/L	(Hammud et al. 2015)
27.	Native rice straw	94.34	Langmuir and Freundlich	Pseudo 1st order	–	6.0	10 h	5.0 g/L	(Gong et al. 2006)
28.	Conch shell powder (CSP)	92.25	Langmuir	Pseudo 2nd order	Endothermic	8.0	120 min	3.0 g/L	(Chowdhury and (Saha) 2011)
29.	20% Phosphoric acid (H_3PO_4) treated + carbonized rice husks [PCP 20]	92.592	Langmuir and Freundlich	–	Endothermic	5.0–6.0	30 min	0.6 g/ 100 mL	(Rahman et al. 2005)
30.	Walnut shell (WS)	90.80	Langmuir	Pseudo 2nd order	Endothermic	5.0	90	0.03 g/ 20 mL	(Dahri et al. 2014)
31.	10% Phosphoric acid (H_3PO_4) treated + carbonized rice husks [PCP 10]	83.33	Langmuir and Freundlich	–	Endothermic	5.0–6.0	30 min	0.8 g/ 100 mL	(Rahman et al. 2005)
32.	White rice husk ash (WRHA)	85.56	Langmuir	Pseudo 2nd order	Exothermic	7.0	120 min	0.45 g/ 100 mL 5.0 g/L	(Tavlieva et al. 2013)
33.	Crude sawdust (CS)	85.47	Langmuir	–	Exothermic	8.0	23 h	2.0 g/L	(Gong et al. 2009)
34.	Bivalve shell-treated Zea mays L. (maize) husk leaf [BS-ZHL]	81.5	Langmuir	Pseudo 1st order	Endothermic	6.0	0.5 h	2.5 g/L	(Jalil et al. 2012)
35.	Maize cob (Zea maize)	80.64	Langmuir and Freundlich	Pseudo 2nd order	–	8.0	25 min	4.0 g/L	(Sonawane and Shrivastava 2009)
36.	Scots Pine (*Pinus silvestris*) sawdust	71.67	Langmuir	Pseudo 2nd order	–	5.0	–	2.0 g/L	(Witek-Krowiak 2013)
37.	*Luffa acutangula* peel	69.64	Langmuir	Pseudo 2nd order	Endothermic	4.0	–	8.0 g/L	(Ng et al. 2015)
38.	Rice bran	68.97	Freundlich	Pseudo 2nd order	Spontaneous and physical	6.0	60–90 min	5.0 g/L	(Wang et al. 2008b)
39.	Formaldehyde-treated sawdust (PCSD)	65.80	–	Pseudo 1st order	–	6.0–10.0	30–45 min	1.0 g/ 100 mL	(Garg et al. 2004)

Table 5 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
40.	Sulphuric acid-treated sawdust (PCSDC)	65.8	–	Pseudo 1st order	–	6.0–10.0	30–45 min	1.0 g/100 mL	(Garg et al. 2004)
41.	Wheat bran	66.57	Freundlich	Pseudo 2nd order	Spontaneous and physical	6.0	60 – 90 min	5.0 g/L	(Wang et al. 2008b)
42.	Lignocellulosic material (TA modified oil palm tree sawdust)	65.79	Langmuir	Pseudo 2nd order	Endothermic	9.0			(Low et al. 2013)
43.	Rattan sawdust (RSD)	62.71	Langmuir	Pseudo 1st order	–	4.0–9.0	210 min	0.30 g	(Hameed and El-Khaiary 2008d)
44.	Eucalyptus bark	59.88	Langmuir	Pseudo 2nd order	Endothermic	5.0	270 min	1.0 g/200 mL	(Boutemedjet and Hamdaoui 2009)
45.	NaOH treated saw dust	58.4795	R–P and Temkin	Pseudo 2nd order	Exothermic	2.9	3 h	4 g/L	(Mane and Babu 2011)
46.	Sodium hydroxide (NaOH) treated + carbonized rice husks [NCP]	57.143	Langmuir	–	Exothermic	5.0 – 6.0	30 min	0.6 g/100 mL	(Rahman et al. 2005)
47.	Eggshell	56.76	Langmuir and Freundlich	Pseudo 2nd order	Endothermic	9.0	90 min	2.0 g/L	(Chowdhury and Das 2012)
48.	Degreased coffee bean	55.3	Langmuir and Freundlich	Pseudo 2nd order	Endothermic	4.0	120 min (2 h)	2.0 g/L	(Baek et al. 2010)
49.	Tamarind seed	54.95	Langmuir	Pseudo 1st order	–	7.0	202 min	2.85 g/L	(Rajeshkannan et al. 2011)
50.	Aerobic granules	56.80	Langmuir	Pseudo 2nd order	Endothermic	Alkaline pH	60 min (50–60 mg/L) 120 min (70–80 mg/L)	0.09 g	(Sun et al. 2008)
51.	*Ananas comosus* (pineapple) leaf powder	54.64	Langmuir	Pseudo 2nd order	Exothermic	9.0	150 min	5.0 g/L	(Chowdhury et al. 2011a)
52.	Hydrothermal carbonization of pine needles (HTC-PN)	52.91	Langmuir	Pseudo 2nd order	Endothermic	7.0	–	2.0 g/L	(Hammud et al. 2015)
53.	Lemon peel	51.73	Langmuir and Redlich–Peterson	–	–	–	24 h	0.5 g/L	(Kumar 2007)
54.	Pine tree root decayed by brown-rot fungi (BRW)	42.63	Langmuir	Pseudo 2nd order	Endothermic	4.0	24 h	3.95 g/L	(Zhang et al. 2011)
55.	Sea shell powder	42.33	Langmuir	Pseudo 2nd order	Exothermic	8.0	120 min	2.0 g/L	(Chowdhury and Saha 2010a)
56.	*Limonia acidissima*(wood apple) shell	34.56	Langmuir	Pseudo 1st order	Endothermic	7.5–8.0	210 min (3.30 h)	0.4 g	(Sartape et al. 2015)

Table 5 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
57.	Jalshakti® (JS), a super absorbent polymer	34.2	Langmuir	Intraparticle Diffusion	–	–	60–90 min	0.8 g/L	(Dhodapkar et al. 2007)
58.	Potato leaves powder, PLP	33.30	Freundlich	Pseudo 2nd order	Exothermic	7.0		2.0 g/L	(Gupta et al. 2014)
59.	Fish (*Labeo rohita*) Scales	38.461	Langmuir	Pseudo 2nd order	Endothermic	8.0	60 min	2.0 g/L	(Chowdhury et al. 2012)
60.	Rubber wood(*Hevea brasiliensis*) sawdust	36.45	Langmuir and Redlich–Peterson	–	–	–	–	–	(Vasanth Kumar and Sivanesan 2007)
61.	Luffa cylindrica	29.40	Langmuir	Pseudo 2nd order	Endothermic	5.0	5 h	0.05 g	(Altınışık et al. 2010)
62.	Potato stem powder (PSP)	27.0	Freundlich	Pseudo 2nd order	Exothermic	7.0		2.0 g/L	(Gupta et al. 2014)
63.	Peroxide treated rice husk (PRH)	26.60	Langmuir	Pseudo 2nd order	Endothermic	8.0	90 min	200 mg/ 100 mL	(Ramaraju et al. 2014)
64.	Hen feathers	26.10	Langmuir	Pseudo 1st order	Endothermic	5.0	150 min	0.1 g	(Mittal 2006)
65.	Marine alga *Caulerpa Racemosa* var. *cylindracea* (CRC)	25.67	Freundlich	Pseudo 2nd order	Endothermic	6.0	300 min	0.1 g/ 25 mL	(Bekçi et al. 2009)
66.	Mesoporous aluminophosphate ($AlPO_4$) molecular sieve	24.51	Langmuir	Pseudo 2nd order	Endothermic	10.0	20 min	0.5 g	(Kannan et al. 2013)
67.	Wheat bran	24.0	Freundlich	–	–	7.0–9.0	40 min	–	(Papinutti et al. 2006)
68.	Sugarcane Bagasse (SB)	23.41	Langmuir	Pseudo 2nd order	Endothermic	6.0	3 h	10 mg	(Xing and Deng 2009)
69.	Nitric acid treated rice husk (NRH)	18.10	Langmuir	Pseudo 2nd order	Endothermic	8.0	90 min	200 mg/ 100 mL	(Ramaraju et al. 2014)
70.	Chemically modified rice husk	17.98	Freundlich	Pseudo 2nd order	Endothermic	7.0	60 min	1.0 g/L	(Chowdhury et al. 2011b)
71.	Rapeseed press cake	17.857	Langmuir	Pseudo 2nd order	Endothermic	6.5	180 min	2.5 mg/L	(Jasińska et al. 2013)
72.	Char from lotus seed biomass	16.75	Langmuir and Freundlich	Pseudo 2nd order	Endothermic	6.84	–	1.5 g/ 150 mL	(Nethaji et al. 2013)
73.	Citric acid (CA) treated pea shells (CAPS)	14.49	Freundlich	Pseudo 2nd order	Endothermic	7.0	35–40 min	6.0 g/L	(Khan et al. 2014)
74.	Waste Pea (*Pisum sativum* L.) shell (PS)	6.20	Freundlich	Pseudo 2nd order	Endothermic	7.0	35–40 min	10.0 g/L	(Khan et al. 2014)

Table 5 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
75.	Sugar cane dust	4.88							(Khattri and Singh 1999)
76.	Neem sawdust	4.354	Langmuir	Pseudo 1st order	Exothermic	7.2	–	–	(Khattri and Singh 2009)
77.	Poultry feathers	3.55	Langmuir	Pseudo 1st order	Endothermic	4.0	120 min	0.2 g/100 mL	(Beak et al. 2009)
78.	Neem saw dust	3.420	Langmuir	–	Exothermic	7.5	–	–	(Khattri and Singh 2000)
79.	Tamarind fruit shell	1.951	Langmuir	Pseudo 2nd order	Endothermic	5.0	60 min	30 g/L	(Saha et al. 2010a)
80.	Mango bark powder (MBP)	0.53 mmol/g	Langmuir	Pseudo 2nd order	Endothermic	6.03	150 min	2.0 g/50 mL	(Srivastava and Rupainwar 2011)
81.	Neem bark powder (NBP)	0.36 mmol/g	Langmuir	Pseudo 2nd order	Endothermic	6.80	120 min	2.0 g/50 mL	(Srivastava and Rupainwar 2011)
82.	Seashell powder	–	–	–	–	–	–	–	(Chowdhury and Saha 2012)
83.	Eggshell powder	–	–	–	–	–	–	–	(Chowdhury and Saha 2012)
84.	Mesoporous borosilicates (MBS)	–	–	–	–	–	–	–	(Paul et al. 2012)
85.	Kapok hull	–	Langmuir	Pseudo 2nd order	Endothermic				(Syed 2011)
86.	Used black tea (UBT)	–	Langmuir	–	Exothermic	–	10 min	0.7 g	(Tahir et al. 2009)
87.	Formaldehyde treated and sulphuric acid treated saw dusts	–	–	–	–	6.9–9.0	120 min	0.4 g/100 mL	(Garg et al. 2003)
88.	NaOH-modified rice husk (NMRH)	–	–	–	–	10.0	–	–	(Chowdhury and Saha 2013)
89.	Sea shell	–	–	Pseudo 2nd order	–	–	–	–	(Chowdhury and Saha 2010b)
90.	Sagaun sawdust	–	Langmuir	–	–	7.5	–	–	(Kattri and Singh 1999)
91.	Pretreated rice husk	–	–	Pseudo 2nd order	–	–	–	–	(Chowdhury and Saha 2011b)
92.	Sugar cane dust	–	Langmuir	–	–	7.5	–	–	(Dadhaniya et al. 2006)
93.	Snail shell (SS)	–							(Oladoja and Aliu 2009)

Table 5 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
94.	Dead leaves of plane tree (*Platanus vulgaris*)	–	Langmuir	Pseudo 2nd order	Endothermic	–	300 min	0.25 g/100 mL	(Hamdaoui et al. 2008b)
95.	De-oiled soya	–	Langmuir	Pseudo 1st order	Endothermic	5.0		0.1 g	(Mittal et al. 2005)
96.	Bottom ash	–			Endothermic	5.0	1–2 h	0.1 g/100 mL	(Gupta et al. 2004)
97.	Ferro-fluid modified sawdust	–	–	–	–	–	–	–	(Safarik et al. 2007)

thermodynamic studies proved that the adsorption was endothermic and chemisorptions ($\Delta H^0 > 40$ kJ/mol) on silica. Recovery of dye on silica was studied from 30 to 90 °C and observed that only 3.5% was recovered from silica. The less recovery on silica proved the strong adsorption of dye on silica.

Talc is the most common silicate material, with an idealized chemical formula of $Mg_3Si_4O_{10}(OH)_2$. Its ability to remove aqueous MG from solution had been studied by Lee et al. (2013).

Zeolites Zeolites are highly porous crystalline aluminosilicates with tetrahedrally connected three dimensional frameworks and extra-framework charge balancing cations. Zeolite structures are commonly described in terms of the size, geometry and connectivity of the pore space. The use of natural zeolite as an adsorbent has gained interest among researchers; mainly because of its sorption properties provide a combination of ion exchange and molecular sieve properties which can also be easily modified (Goursot et al. 1997; Sánchez and Pariente 2011).

Zeolites have been used as chemical sieves, as water softeners, as removers of ammonia from urban, agricultural and industrial waste waters and of toxic gases from gaseous emissions, as filters for odor control, as chemical fertilizer, herbicide and pesticide carriers and in animal nutrition (Sand and Mumpton 1978; Breck 1984; Karge and Weitkamp 1989).

Han et al. (2010) investigated the use of natural zeolite, obtained from Xinyang city in China, for the removal of MG from aqueous solution. The rate constants of sorption for different kinetic models were calculated. Good correlation coefficients were obtained for the pseudo-second order kinetic model. In addition, authors were treated spent zeolite by microwave irradiation and it was found that yield of regeneration was 85.8% in the case of microwave irradiated time 10 min at 160 W.

Table 7 represented the maximum adsorption capacities together with the isotherm, kinetics and thermodynamic results of various natural materials used for the removal of MG

Biosorbents

Biosorption is defined as the accumulation and concentration of pollutants from aqueous solutions by the use of biological materials. Biological materials, such as yeast, fungi or bacterial biomass and biopolymers are used as adsorbents for the removal of MG from aqueous media (Raval et al. 2016d; Gupta et al. 2015).

Biomass Biomass is biological material derived from living, or recently living organisms, which can be applied

Table 6 The maximum adsorption capacities and other optimal conditions of various industrial waste materials for adsorption of MG from water and wastewater

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
1.	Acid-activated sintering process red mud (ASRM)	336.40	Langmuir	Pseudo 2nd order	Endothermic	>3.2	180 min	0.25 g/ 250 mL	(Zhang et al. 2014b)
2.	Bagasse fly ash	170.33	Freundlich	Pseudo 2nd order	–	7.0	240 min	1 g/L	(Mall et al. 2005)
3.	Activated Slag	74.16 (batch capacity) 78.00 (column capacity)	Langmuir and Freundlich	Pseudo 1st order	Endothermic	10.0	6–8 h	1.0 g/L	(Gupta et al. 1997)
4.	Fly ash	40.65	Langmuir	Pseudo 2nd order	Endothermic	>6.0	–	2.0 g/L	(Witek-Krowiak et al. 2012)
5.	$Ca(OH)_2$ –Treated Fly Ash	17.383	Langmuir	Pseudo 2nd order	Exothermic	7.0	120 min	1.0 g	(Chowdhury and Saha 2011a)
6.	Bottom ash	6.2×10^{-3}	Langmuir	Pseudo 2nd order	Endothermic	5.0	4500 s	0.1 g	(Gupta et al. 2004)
7.	Carbon based adsorbents [waste material from paper industry (WMPI) and pine bark (PB)]	–	–	–	–	–	–	0.05 mg/ 20 mL	(Méndez et al. 2007)

to both animal and vegetable derived material. The major advantages of biosorption over conventional treatment methods include, low cost; high efficiency; minimization of chemical or biological sludge; no additional nutrient requirement and possibility of regeneration of biosorbent (Volesky 1990).

Decolorization/bioadsorption of MG dye from wastewater by (dead/living) biomass of submerged aquatic plant (Rajesh Kannan et al. 2010); white-rot fungi (Kuhar and Papinutti 2013); fresh water algae (Kumar et al. 2005); brown marine algae (Kannan et al. 2009) and immobilized baker's yeast cells (Godbole and Sawant 2006) were studied by many researchers (shown in Table 8)

Wang (2009) used alligator weed (a freshwater macrophyte) as biosorbent for the removal of basic dye malachite green from aqueous solution mainly because alligator weed, originated from South America, occupies agricultural areas and blocks drainage and irrigation channels causing problems on agricultural land. In addition, it causes water pollution from plant decomposition and an increase in mosquito breeding areas. Finally, author concluded that the alligator weed could serve as low-cost adsorbents for removing MG from aqueous medium.

Parthenium (*Parthenium hysterophorus* L.), also known as white top or carrot weed, an annual herbaceous weed was pretreated with H_3PO_4 and H_2SO_4 by Lata et al. (2008) and employed for the adsorption of MG dye.

Biopolymers Biopolymers are high molecular weight compounds synthesized by living organisms. The performance of chitin hydrogel as an adsorbent to remove MG has been demonstrated by Tang et al. (2012). They found that the maximum adsorption capacity of hydrogel for MG was 26.88 mg/g. Authors concluded from the calculated activation energy (37.18 kJ/mol) and from the results of SEM and FTIR that the adsorption of malachite green on the hydrogel was physical process.

Bekçi et al. (2008) synthesized chitosan beads and examined its adsorption capacity towards MG. In contrast to the chitin hydrogels the authors concluded from the calculated activation energy value (85.6 kJ/mol) that the adsorption of MG onto chitosan beads was chemical in nature.

Although chitin and chitosan are abundantly available biopolymers and are biocompatible and biodegradable material, they have less chemical versatility due to their strong crystal structure, and hence their chemical structures

Table 7 The maximum adsorption capacities and other optimal conditions of various natural materials for adsorption of MG from aqueous solutions

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	Reference
1.	Surfactant-modified alumina (SMA)	185.00	Langmuir	–	–	–	1 h	1.0 g/L	(Das et al. 2009a)
2.	Sodium dodecyl sulfate (SDS) modified alumina	185.00	Langmuir	–	–	–	1 h	1.0 g/L	(Das et al. 2009b)
3.	Bentonite	178.60	Langmuir	Pseudo 2nd order	Endothermic	5.0	60 min	1 g/L	(Bulut et al. 2008)
4.	Aminopropyl functionalized magnesium phyllosilicate (AMP) clay	130.64	Langmuir	Pseudo 1st order	Endothermic	9.8	180 min	0.1 mg/mL	(Lee et al. 2011)
5.	Clayey soil of Indian origin	78.57	Langmuir	Pseudo 2nd order	Exothermic	6.0	90 min	3 g/L	(Saha et al. 2010b)
6.	Kaolin	65.42	Langmuir	Pseudo 2nd order	Exothermic	7.0	90 min	2 g/L	(Nandi et al. 2009)
7.	Non-tetrahedral alumina	61.35	Langmuir	–	Endothermic	5.0	4 h	250 mg/50 mL	(Kannan et al. 2008)
8.	Organically modified clay	56.82	Langmuir	Pseudo 2nd order	Endothermic	6.0	30 min		(Arellano-Cárdenas et al. 2013)
9.	Low-cost inorganic powder (Persian Kaolin)	52.91	Langmuir	Pseudo 2nd order	Endothermic	7.0	5 min	0.8 g/L	(Tehrani-Bagha et al. 2011)
10.	Bentonite clay	7.716	Langmuir, Freundlich and D–R	Pseudo 1st order	Endothermic	9.0	10 min	0.05 g	(Tahir and Rauf 2006)
11.	Surfactant-Coated Alumina (SDS-Al2O3)	2.78	–	–	–	4.7	–	500 mg	(Farhadi et al. 2010)
12.	Rarasaponin–bentonite	0.93 mmol/g	Langmuir	–	–	8.0	2 h	–	(Kurniawan et al. 2012)
13.	Natural kaolinitic-clay	0.64 mmol/g	Langmuir	–	Exothermic	7.0	1 day	1.0 g/L	(Issa et al. 2014)
14.	Iron humate (IH)	0.0527 mmol/g	Langmuir	–	–	–	–	–	(Janoš 2003)
15.	Natural rarasaponin modified kaolin	–	Sips	Pseudo 1st order	Exothermic	–	–	–	(Suwandi et al. 2012)
16.	Inorganic–organic pillared clay granules	–	Langmuir	Pseudo 1st order	–	6.0	120 min	–	(Cheknane et al. 2010)
17.	Montmorillonite	–	Langmuir	–	Endothermic	–	–	0.1 g	(Tahir et al. 2010)
18.	Organomineral sorbent–iron humate	–	–	Pseudo 2nd order	–	–	–	–	(Janoš and Šmídová 2005)
19.	Bentonite	–	Temkin and Redlich-Peterson isotherms	–	–	8.2	60 min	1.0 g	(Özdemir and Keskin 2009)

Table 7 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	Reference
20.	Activated α-alumina	–	–	–	–	10.0	10 min	0.1 g/ 250 mL	(Rogaieh and Gharbani 2013)
21.	Sand	–	Freundlich	Pseudo 2nd order	–	4.8	–	0.03 g	(Rauf et al. 2008)
22.	Zeolite	46.35	Langmuir	Pseudo 1st order	–	6.0	–	–	(Wang and Ariyanto 2007)
23.	Tetrahedral silica	45.05	Langmuir	–	Endothermic	6.0	5 h	150 mg/ 50 mL	(Kannan et al. 2008)
24.	Natural zeolite	23.55	–	–	–	–	–	–	(Wu et al. 2010)
25.	Talc (silicate material)	20.968	Langmuir	Pseudo 2nd order	–	8.0	–	–	(Lee et al. 2013)
26.	Perlite	3.36	Freundlich	Pseudo 1st order	Exothermic	8.0–9.0	40 min	0.1 g/ 50 mL	(Govindasamy et al. 2009)
27.	Philipsite-rich-zeolite tuff	1.22 mmol/g	Langmuir	–	Exothermic	7.0	1 day	1.0 g/L	(Issa et al. 2014)
28.	Natural zeolite	–	Koble–Corrigan model	Pseudo 2nd order	Endothermic	4.0	480 min	0.03 g/ 10 mL	(Han et al. 2010)
29.	Silica gel	–	–	–	–	–	–	–	(Samiey and Toosi 2010)

have been modified to overcome this undesirable characteristic. In an effort to increase the chemical versatility and adsorption efficiency, chitin and chitosan biopolymers have been modified or crosslinked by various surfactants and crosslinking agents.

Huang et al. (2012) explored the use of Chitin grafted poly (acrylic acid) (chi-g-PAA) for the removal of MG from wastewater and concluded that the carboxylic groups that are grafted to the surface of chitin can be hydrolyzed to a sodium salt using dilute $NaOH_{(aq)}$ and can be readily utilized to adsorb malachite green from aqueous solution. The monolayer adsorption capacity obtained was 285.70 mg/g which was 10 times higher than the chitin hydrogels (26.88 mg/g).

Wan Ngah et al. (2010) studied the removal of MG from aqueous solutions through adsorption on to chitosan coated bentonite beads. The characterization of adsorbents was performed by Fourier Transform Infrared (FTIR) spectroscopy, scanning electron microscopy (SEM) coupled with energy dispersive spectroscopy (EDS) and X-ray diffraction (XRD) analysis. Authors mentioned that the monolayer adsorption capacity obtained was 435.00 mg/g which was five times greater than the chitosan beads (93.55 mg/g).

Peat Peat is a porous and complex natural material, widely available and studied as an alternative adsorbent for different pollutants as well as dyes (Raval et al. 2016c).

Coco-peat was used by Vijayaraghavan et al. (2015) as an adsorbent to study the adsorption of malachite green and crystal violet from wastewater. It was found that a contact time of 60 min was sufficient for the adsorption to reach equilibrium. The optimum pH was found to be 7.0. The data for the adsorption of MG was described well by both the Sips adsorption isotherm model. The adsorption process was found to be endothermic.

Nanomaterials

Typical nanomaterials include nanotubes, nanoparticles, nanofibers, nanorods, fullerenes, and nanowires (Schmidt et al. 2002; Gupta and Saleh 2013; Gupta and Nayak 2012; Saravanan et al. 2013, 2014).

Carbon nanotubes were discovered accidentally by Iijima (1991), while studying the surfaces of graphite electrodes used in an electric arc discharge. CNTs are divided into two types based on the arrangement of their graphene cylinders: (1) single-walled CNTs (SWCNTs)

Table 8 The maximum adsorption capacities and other optimal conditions of various biosorbents for adsorption of MG from water and wastewater

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
1.	Freshwater macrophyte alligator weed	185.54	Langmuir	Ritchie second-order	–	6.0	–	7.0 g/L	(Wang 2009)
2.	*Firmiana simplex* wood fiber	>142.40	Freundlich	Pseudo 2nd order	Endothermic	11.0	180 min	0.25 g	(Pan and Zhang 2009)
3.	*Pleurotus ostreatus* (A macro-fungus)	125.00	Freundlich	Pseudo 2nd order	–	8.0	240 min	4.0 g/L	(Chen et al. 2014)
4.	*Pithophora* sp., a fresh water algae	117.65	Redlich-Peterson	Pseudo 2nd order	–	5.0	8 h	0.015 g	(Kumar et al. 2005)
5.	*Hydrilla verticillata* biomass	91.97	Freundlich	Pseudo 2nd order	–	8.0	200 min	0.26 g	(Rajeshkannan et al. 2010)
6.	Ashoka (*Saraca asoca*) leaf powder (ALP)	83.30	Freundlich	Pseudo 2nd order	Exothermic	6.0	25 min	2.0 g/L	(Gupta et al. 2012)
7.	*Hydrilla Verticillata* Biomass	69.88	Langmuir	Pseudo 1st order	–	8.0	150 min	0.55 g	(Rajesh Kannan et al. 2010)
8.	*Turbinaria conoides* brown marine algae	66.60	Langmuir	Pseudo 2nd order	–	8.0	150 min	0.55 g	(Kannan et al. 2009)
9.	Native anaerobic granular sludge	61.73	Langmuir	Pseudo 2nd order	Exothermic	5.0	2 h	2.4 g/L	(Cheng et al. 2008)
10.	Heat-treated anaerobic granular sludge	59.17	Langmuir	Pseudo 2nd order	Exothermic	5.0	2 h	2.4 g/L	(Cheng et al. 2008)
11.	Eriobotrya japonica (Thunb.) Lindley (Loquat) seed biomass	57.47	Langmuir	Pseudo 2nd order	Endothermic	5.0	60 min	10.0 g/L	(Aksakal et al. 2009)
12.	*Daucus carota* plant (carrot leaves powder: CLP)	52.60	Freundlich	Pseudo 2nd order	Exothermic	7.0	30 min	2.0 g/L	(Kushwaha et al. 2014)
13.	Mango (*Mangifera indica* L.) seed husks [MSH]	47.90	Freundlich	Pseudo 2nd order	Exothermic	5.0	2.0 h	5.0 g/L	(Franca et al. 2010b)
14.	*Daucus carota* plant (carrot stem powder: CSP)	43.40	Freundlich	Pseudo 2nd order	Exothermic	7.0	30 min	2.0 g/L	(Kushwaha et al. 2014)
15.	Dead needles of Aleppo pine (*Pinus halepensis*)	33.56	Freundlich	Intraparticle diffusion	Endothermic	–	330 min	0.5 g/ 300 mL	(Hamdaoui et al. 2008a)
16.	Almond Shell (*Prunus dulcis*)	29.0	Langmuir	Pseudo 2nd order	Endothermic	–	180 min	20 g/L	(Ozdes et al. 2010)
17.	Phosphoric acid treated Parthenium hysterophorus Linn. Biomass (PWC)	25.00	Langmuir	Pseudo 2nd order	–	7.0	90 min	0.2 g/ 50 mL	(Lata et al. 2008)
18.	Microwave assisted zinc chloride activated epicarp of *Ricinus communis* (ZRC)	24.39	Langmuir	Pseudo 2nd order	–	5.0		0.2 g/mL	(Makeswari and Santhi 2013)
19.	Immobilized *Saccharomyces cerevisiae*	17.0	Freundlich	–	–	–	–	–	(Godbole and Sawant 2006)
20.	Sulfuric acid treated Parthenium hysterophorus Linn. Biomass	16.81	Langmuir	Pseudo 2nd order	–	7.0	90 min	0.2 g/ 50 mL	(Lata et al. 2008)
21.	Microwave activated epicarp of *Ricinus communis* (MRC)	12.65	Langmuir	Pseudo 2nd order	–	5.0	–	0.2 g/mL	(Makeswari and Santhi 2013)

Table 8 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
22.	*Hydrilla verticillata* biomass	–	–	–	–	8.4	194.50 min	11.14 g/L	(Rajeshkannan et al. 2009)
23.	Pithophora sp., a fresh water algae	–	–	Pseudo 1st order	–	6.0	–	–	(Vasanth Kumar et al. 2006)
24.	Yeast *S. cerevisiae* MTCC 174	–	–	Pseudo 2nd order	Endothermic	6.88	60 min	0.49 g	(Singh et al. 2012)
25.	Chlorella-based biomass	–	–	Pseudo 2nd order	–	7.0		2.0 g/ 2.0 dm^3	(Tsai and Chen 2010)
26.	White-rot fungus *Ganoderma lucidum*	–	–	–	–	–	–	–	(Kuhar and Papinutti 2013)
27.	Carboxymethyl cellulose-acrylic acid	555.56	Temkin	Pseudo 2nd order	Endothermic	–	120 min (1 h)	–	(Zhang et al. 2014a)
28.	Chitosan coated bentonite (CCB) beads	435.0	Freundlich	Pseudo 2nd order	–	6.0	30 min	0.01 g/ 50 mL	(Wan Ngah et al. 2010)
29.	Cellulose modified with maleic anhydride (CMA)	370.00	Langmuir	Pseudo 2nd order	Exothermic	–	–	–	(Zhou et al. 2015)
30.	Amylopectin and poly (acrylic acid) (AP-g-PAA)	352.11	Langmuir	Pseudo 2nd order	Endothermic	7.0	30 min	50 mg	(Sarkar et al. 2014)
31.	Chitin grafted poly (acrylic acid)	285.70	Langmuir	–	–	–	–	–	(Huang et al. 2012)
32.	Chitosan beads	93.55	Langmuir	Pseudo 2nd order	Endothermic	8.0	300 min (5 h)	10 g/L	(Bekçi et al. 2008)
33.	Cyclodextrin-based adsorbent	91.90	Langmuir	Pseudo 2nd order	–	8.0	120 min	0.15 g	(Crini et al. 2007)
34.	Lignin sulfonate polymer	70.077	Langmuir	Pseudo 2nd order	Endothermic	7.0	300 min	0.05 g	(Tang et al. 2014)
35.	Bio-polymer (Cellulose)	2.422	Langmuir and Freundlich	Pseudo 2nd order	Exothermic	7.2	30 min (0.5 h)	5 g/L	(Pradeep Sekhar et al. 2009)
36.	Chitin hydrogels	26.883	Langmuir	Pseudo 2nd order	Endothermic	7.0	25 h	–	(Tang et al. 2012)
37.	Coco-peat	276.80	Sips	Pseudo 1st order	Endothermic	7.0	60 min	0.2 g/ 100 mL	(Vijayaraghavan et al. 2015)
38.	Peat of Brunei Darussalam (Keramut peat)	0.31 mmol/ g	Redlich–Peterson (R–P) model	Pseudo 2nd order	Endothermic	6.0	4 h	0.050 g	(Chieng et al. 2014)

and (2) multi-walled CNTs (MWCNTs) (Khani et al. 2010; Saleh and Gupta 2011).

MWNTs are collections of several concentric graphene cylinders and are larger structures compared to SWNTs which are individual cylinders of 1–2 nm diameter. The former can be considered as a mesoscale graphite system, whereas the latter is truly a single large molecule (Odom et al. 1998). The mechanisms by which the dye molecules are sorbed onto CNTs are very complicated and appear attributable to electrostatic attraction, sorption, precipitation and chemical interaction between the reactive groups of dye molecules and the surface functional groups of CNTs.

Recently, Rajabi et al. (2016) functionalized MWCNTs with the carboxylate group and used as an adsorbent for the rapid and fast removal of MG dye from the aqueous solutions. The optimized contact time and pH were 10 min and 9, respectively. The effect of temperature revealed that the adsorption capacity of MG dye increased with increasing contact time, temperature and pH. The

Table 9 The maximum adsorption capacities and other optimal conditions of various nanomaterials, composites and nanocomposite adsorbents for the removal of MG from water and wastewater

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
1.	Copper nanowires loaded on activated carbon	434.80	Langmuir	Pseudo 2nd order	–	5.0	20 min	0.1 g	(Ghaedi et al. 2015)
2.	Zinc oxide nanoparticle loaded on activated carbon (ZnO-NP-AC)	322.58	Langmuir	Pseudo 2nd order	–	7.0	15 min	0.005 g	(Ghaedi et al. 2014)
3.	Metal oxide nanoparticles (ZnO)	310.50	Langmuir and Freundlich	Pseudo 2nd order	Endothermic	8.0	120 min		(Kumar et al. 2013)
4.	Maghemite nanoparticles (γ-Fe_2O_3)	227.30	Langmuir	–	–	3.0	–	0.10 g	(Afkhami et al. 2010)
5.	Metal oxide nanoparticles (SnO_2)	216.90	Langmuir and Freundlich	Pseudo 2nd order	Endothermic	8.0	120 min		(Kumar et al. 2013)
6.	Gold nanoparticles loaded on activated carbon (Au-NP-AC)	164.57	Langmuir	Pseudo 2nd order	Endothermic	7.0	4.4 min	0.015 g	(Roosta et al. 2014)
7.	*N*-Benzyl-*O*-carboxymethylchitosan magnetic nanoparticles (OC-BzM)	144.79	Langmuir and Freundlich	Pseudo 2nd order	Endothermic	7.0	60 min	25 mg	(Debrassi et al. 2012)
8.	Functionalized multi wall carbon nanotubes (*f*-MWCNTs)	142.85	Langmuir	Pseudo 2nd order	–	7.0	80 min	0.08 g/ 100 mL	(Shirmardi et al. 2013)
9.	NiO nano flake-based flowerlike architectures	142.08	Langmuir	Pseudo 2nd order	–			333.3 mg/ L	(Wei et al. 2014)
10.	Silver nano particles (AgOH-NP) loaded on activated carbon prepared from *Rosa canina* tree (AgOH-NP-AC-RC)	105.00	Freundlich	Pseudo 2nd order	–	8.0	20 min	0.09 g/ 50 mL	(Tavakol et al. 2015)
11.	Superparamagnetic sodium alginate-coated Fe_3O_4 nanoparticles (Alg–Fe_3O_4)	47.84	Langmuir	Pseudo 2nd order	Endothermic	9.0	20 min	0.1 g/ 50 mL	(Mohammadi et al. 2014)
12.	Cadmium hydroxide nanowires loaded on activated carbon ($Cd(OH)_2$-NW-AC)	19.0	Langmuir	Pseudo 2nd order	Endothermic	–	–	–	(Ghaedi and Mosallanejad 2014)
13.	Melamine supported magnetic iron oxide nanoparticles (Fe_3O_4@Mel)	9.06	Freundlich	Pseudo 2nd order	–	6.5	240 min	0.3 g	(Mirzajani and Ahmadi 2015)
14.	Citrate stabilized magnetite nanoparticles	0.704	Freundlich	Pseudo 2nd order	–	4.0	–	10 mg	(Mishra et al. 2014)
15.	$FeCl_2$/$FeCl_3$/perlite magnetic nanoparticles ($FeCl_2$/ $FeCl_3$/PMNs),	0.01	BET	Pseudo 2nd order	–	6.0	120 min	0.05 g	(Heydartaemeh et al. 2014)
16.	MnO_2 nanosheets	–	–	Pseudo 1st order	–	5.6	120 min	0.5 g/L	(Saha and Pal 2014)
17.	Green tea extract based iron nanoparticles (GT-Fe NPs)	–	–	Pseudo 1st order	–	9.0	–	1.12 g/L	(Weng et al. 2013)

Table 9 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
18.	TiO_2 nanoparticles	–	–	Pseudo 2nd order	–	6.9	–	3.0 g/L	(Suresh and Annadurai 2013)
19.	Iron nanoparticles (Fe NPs)	–	–	–	–	–	–	–	(Huang et al. 2014)
20.	Reduced graphene oxide (RGO)–Fe_3O_4nanoparticles (NPs)	–	–	–	–	–	–	–	(Geng et al. 2012)
21.	Halloysite nanotubes (HNTs)	99.60	Langmuir	Pseudo 2nd order	Endothermic	9.5	30 min	0.2 g	(Kiani et al. 2011)
22.	ZnS	98.3%	–	–	–	8.0	120 min	0.08 g/L	(Rajabi et al. 2013)
23.	ZnS:Fe	99.0%	–	–	–	8.0	120 min	0.08 g/L	(Rajabi et al. 2013)
24.	Fe3O4@SiO2-NH2	–	Freundlich	Pseudo 2nd order	–	7.0	–	100 mg/L	(Sun et al. 2015b)
25.	Lac-PAM-CTS	–	–	–	–	–	–	–	(Sun et al. 2015a)
26.	Magnetic β-cyclodextrin-graphene oxide nanocomposites(Fe_3O_4/β-CD/GO)	990.10	Langmuir	Pseudo 2nd order	Endothermic	7.0	120 min	–	(Wang et al. 2015)
27.	K-Carrageenan-g-AA	833.33	Langmuir	Pseudo 2nd order	Exothermic	7.0	180 min	0.05 g	(Pourjavadi et al. 2014)
28.	Kappa-carrageenan-g-polyacrylic acid/TiO_2–NH_2 hydrogel nanocomposite (K-Carrageenan-g-AA/TiO_2–NH_2)	666.66	Langmuir	Pseudo 2nd order	Exothermic	7.0	180 min	0.05 g	(Pourjavadi et al. 2014)
29.	Melamine/maleic anhydride (S-Me/MA)	641.03	Langmuir	Pseudo 2nd order	Endothermic	9.0	24 h	0.06 g	(Rong et al. 2014)
30.	Iron nanoparticles loaded on ash (nFe-A)	500	Langmuir	Pseudo 2nd order	Endothermic	8.0	30 min	–	(Agarwal et al. 2016)
31.	Poly(vinyl alcohol) (PVA) and chitosan (CS)(PVA/CS composite)	380.65	Freundlich	Pseudo 2nd order	Endothermic	6.0	540 min	0.1 g/ 50 mL	(Li et al. 2012)
32.	Alginate/polyaspartate composite hydrogel beads	300–350	–	–	–	–	3.0 h	0.1 g/ 50 mL	(Jeon et al. 2008)
33.	Polyacrylic acid (PAA)–nanoclay nanocomposite	243.11	Freundlich	–	–	9.2	15 min	0.5 g/ 100 mL	(Sonawane et al. 2009)
34.	Fe_3O_4@PANI nanocomposites	240.00	Langmuir	Pseudo 2nd order	Exothermic	7.0	4 h	0.01 g	(Mahto et al. 2014)
35.	Vinyl-modified mesoporous poly(acrylic acid)/SiO_2 (PAA/SiO_2) composite nanofiber membranes	220.49	Redlich–Peterson	Pseudo 2nd order	–	–	240 min	50 mg/ 50 mL	(Xu et al. 2012)
36.	Activated carbon/$CoFe_2O_4$ composite (AC/CFO)	89.29	Langmuir	Pseudo 2nd order	–	5.0	5 min	–	(Ai et al. 2010)

Table 9 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
37.	Unsaturated polyester Ce(IV) phosphate (USPECe(IV)P) composite	44.85	Freundlich	Pseudo 2nd order	Endothermic	8.0	30–35 min	0.1 g/ 100 mL	(Khan et al. 2013)
38.	Graphene oxide (GO)/cellulose bead (GOCB) composites	30.090	Langmuir	Pseudo 2nd order	Endothermic	7.0	12 h	0.1 g	(Zhang et al. 2015)
39.	Carbon nanotube/polyaniline composites	15.45	Langmuir	Pseudo 2nd order	Endothermic	–	–	–	(Zeng et al. 2013)
40.	MWCNTs functionalized with the carboxylate group	11.73	Langmuir	Pseudo 2nd order	Endothermic	9.0	10 min	–	(Rajabi et al. 2016)
41.	Magnetoactive nanocomposite fibers	8.809	Langmuir	–	Endothermic	8.0	24 h	–	(Savva et al. 2015)
42.	Polyaniline–nickel ferrite magnetic nanocomposite	4.09	Langmuir	Pseudo 2nd order	–	7.0	210 min	5.0 g/L	(Patil and Shrivastava 2014)
43.	Humic acid-immobilized amine modified polyacrylamide/bentonite composite (HA-Am-PAA-B)	656.50 (μmol/g)	Freundlich	Pseudo 2nd order	–	8.0	3 h	2.0 g/L	(Anirudhan and Suchithra 2009)
44.	Humic acid-immobilized amine-modified polyacrylamide-bentonite composite (HA-Am-PAA-B)	656.50 (μmol/g)	Freundlich	Pseudo 2nd order	–	5.0–8.0	1 h	2.0 g/L	(Anirudhan et al. 2009)
45.	Glycine–modified silica nanoparticles	–	Langmuir	Pseudo 1st order	–	10.0	40 min	–	(Mansa et al. 2016)
46.	Magnetic *Pycnoporus sanguineus*-Loaded Alginate Composite Beads	–	–	–	–	–	–	–	(Yang et al. 2014)
47.	Polyaniline/ZnO nanocomposite	–	–	–	–	–	5 h	0.4 g/L	(Eskizeybek et al. 2012)
48.	$Fe_{0.01}Ni_{0.01}Zn_{0.98}O$/ polyacrylamide nanocomposite	–	–	–	–	–	–	–	(Kant et al. 2014)
49.	Hierarchically macro/micro-porous silicon–carbon–nitrogen (Si–C–N) hybrid material	–	–	–	–	–	–	–	(Meng et al. 2015)

adsorption equilibrium and kinetic data was well fitted and found to be in good agreement with the Langmuir isotherm model and pseudo second order kinetic model, respectively.

Shirmardi et al. (2013) prepared functionalized multi-walled carbon nanotubes (*f*-MWCNTs) by acid treatment and used for the adsorption of MG dye from aqueous solution in batch mode. The results indicated that by increasing contact time, pH and adsorbent dosage, the removal percentage increased, but by increasing the initial MG concentration, the removal percentage decreased. Based on correlation coefficient (R^2), the experimental data follows the Langmuir isotherm with maximum adsorption capacity of 142.85 mg/g.

Halloysite, a mineral of the kaolin group [forms hollow tubular crystals (halloysite nanotubes—HNTs)], can be used as MG dye adsorbent by Kiani et al. (2011) and found that the equilibrium adsorption was reached within 30 min. The adsorption of MG by HNTs obeyed pseudo-second-order kinetics with activation energy 18.28 kJ/mol,

suggested that the process was physisorption. The best-fit adsorption isotherm was achieved with the Langmuir model, indicating that homogeneous adsorption occurred. The negative values of ΔG° and positive value of ΔH° showed that the adsorption was a spontaneous and endothermic.

The particles of at least one dimension smaller than 1 μm, and potentially as small as atomic and molecular length scales (0.2 nm) are called nanoparticles. Nanoparticles can have amorphous or crystalline form (Buzea et al. 2007). Due to some of the unique characteristics such as small size, catalytic potential, large surface area, ease of separation, and large number of active sites for interaction with different contaminants the nanoparticles have established themselves as excellent adsorbents (Raval et al. 2016d).

Composites and nano-composites

The nano-scale inclusions that are imbedded within matrix of a material are referred as composites (Chen et al. 2013).

Poly(vinyl alcohol) (PVA) is a water-soluble material containing many reactive hydroxyl groups. It has been extensively applied in biomedical and pharmaceutical fields due to its low cost, non-toxicity, biocompatibility, good mechanical strength and chemical stability. However, the PVA foam illustrated low adsorption for organic and inorganic pollutants. Chitosan (CS) is one of the most abundant biopolymers in nature. It exhibits good adsorption capacity for organic dyes and heavy metals because it has a large number of hydroxyl and amine groups. However, the use of CS in the form of flake or powder cannot be separated easily. Therefore, Li et al. (2012) prepared a novel foam adsorbent based on PVA/CS composites by the interpenetrating polymer network (IPN), foaming process and freeze-thaw treatment and found that the composite material had higher adsorption efficiency (380.65 mg/g) compared with the single component.

Hussain et al. (2006) defined nanocomposites as materials consisting of at least two phases with one dispersed in another that is called matrix, and thus forms a three dimensional network.

Wang et al. (2015) prepared Magnetic β-cyclodextrin-graphene oxide nanocomposites (Fe_3O_4/β–CD/GO) as an adsorbent for MG dye because graphene oxide have abundant hydrophilic groups on its surface, and thus it has good adsorption capacity still its separation was difficult task from sample solution and hence, to simplify the retrieval procedure, magnetic adsorbent (Fe_3O_4) has been incorporated with GO. In addition, β-CD was also added onto composite (Magnetic-graphene oxide) to provide hydrophobic inner cavity and a hydrophilic exterior which can enhance the adsorption capacity of composite adsorbent for MG dye.

Table 9 presented the maximum adsorption capacities of various nanomaterials, composite and nanocomposite adsorbents used for MG.

Miscellaneous adsorbents

See Table 10.

Table 10 The maximum adsorption capacities and other optimal conditions of various miscellaneous adsorbents for the removal of MG from water and wastewater

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
1.	Zeolitic imidazole framework-67 (ZIF-67)	3227.00	Langmuir	Pseudo 2nd order	Endothermic	–	2 h	0.25 g/L	(Lin and Chang 2015)
2.	Graphite oxide (GO)	248.00	Langmuir	Pseudo 2nd order	–	–	20 min	10 mg/ 500 mL	(Bradder et al. 2011)
3.	Poly-γ-glutamic acid (γ-PGA)	293.32	Redlich-Peterson	Pseudo 2nd order		>5.0	2 h	0.4 g/L	(Stephen Inbaraj et al. 2006)
4.	Defective coffee beans press cake	208.30	Langmuir	Pseudo 2nd order	–	5.0	4.0 h	5.0 g/L	(Franca et al. 2010a)
5.	Polyethylene terephthalate (PET) by chemical activation with NaOH (PETNa8)	169.49	Langmuir	Pseudo 1st order	–	10.0	90 min	0.1 g/ 25 mL	(Akmil-Başar et al. 2005)
6.	Magnetic sorbent (“blue magnetite”)	93.20	Langmuir	–	–	–	30 min	–	(Šafařík et al. 1995)

Table 10 continued

Sr. No.	Adsorbents	Q_{max} (mg/g)	Isotherm study	Kinetics study	Thermodynamic study	pH	Equilibrium time	Dosage of adsorbent	References
7.	Polymeric gel	4.900	Langmuir	Pseudo 2nd order	Exothermic	–	–	–	(Malana et al. 2010)
8.	Polylactide (PLA)/spent brewery grains (SBGs) films	1.484	Freundlich	Pseudo 2nd order	–	6.89	95 min	0.42 g	(Chanzu et al. 2012)
9.	Polyethylene glycol (PEG) micelles	–	Langmuir	–	Endothermic	–	10 min	–	(Chen et al. 2009)
10.	polystyrene microspheres	–	–	–	–	–	–	–	(Eckenrode et al. 2005)
11.	Magnetic affinity adsorbent (blue magnetite)	–	–	–	–	–	–	–	(Šafařík and Šafaříková 2002)
12.	Loosely bound extracellular polymeric substances (LB-EPS), tightly bound EPS (TB-EPS)	–	–	–	–	7.0	–	–	(Gao et al. 2011)
13.	Amberlite XAD-4 resin	–	–	Pseudo 1st order	Endothermic	–	150 min	–	(Dos Reis et al. 2011)
14.	AmberliteXAD-2 resin	–	–	Pseudo 1st order	Endothermic	–	150 min	–	(Dos Reis et al. 2011)
15.	Metal–organic framework MIL-100(Fe)	–	Freundlich	Pseudo 2nd order	Endothermic	5.0	–	–	(Huo and Yan 2012)
16.	D201_Lac	–	–	–	–	–	–	–	(Zhang et al. 2012)
17.	Mesoporous Cu/Al_2O_3–MCM-41	–	–	–	–	11.0	30 min	1.0 g/L	(Pradhan et al. 2011)
18.	Calcium oxalate—bromopyrogallol red inclusion sorbent	–	–	–	–	–	–	–	(Wang and Gao 2009)
19.	Polyurethane foam (PUF) loaded with sodium dodecylsulfate (SDS)	–	–	–	–	–	–	–	(Robaina et al. 2009)
20.	Functionalized magnetic adsorbents (FMAs)	–	–	–	–	–	–	–	(Wang et al. 2014b)

Conclusions and future perspectives

Adsorption process being very easy, economical, effective and versatile had become the most favored methods for amputation of toxic and hazardous dyes from wastewater. This review article revealed about various adsorbents, such as activated carbons, low-cost materials, nanomaterials, composites and nanocomposites, used for the adsorption and removal of MG from water and wastewater. It was concluded that amongst all the other adsorbents, agricultural solid wastes and biosorbents, such as biopolymers and biomass, have been extensively used for the removal of MG and are acknowledged as effective and economic adsorbents for the removal of MG dye. Various experimental conditions (pH, temperature, contact time and amount of adsorbent) have been also mentioned for related adsorbents in the present article.

This review article also disclosed that the Langmuir and Freundlich adsorption isotherm models were frequently applied to evaluate the adsorption capacity of various adsorbents towards MG. In kinetic study, it was observed that the pseudo-second-order rate model was been followed for the adsorption of MG and according to thermodynamic parameters the adsorption process was found to be spontaneous in nature.

Since, most of the reported studies had been performed using batch and small-scale column adsorption tests, further research is required for the development of more effective adsorbents, modeling of adsorption mechanism, regeneration of spent adsorbents and treatment of real industrial wastewater.

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